This Page Is Inserted by IFW Operations and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problem Mailbox.

THIS PAGE BLANK (USPTO)





(1) Publication number: 0 554 015 A1

12)

EUROPEAN PATENT APPLICATION

(21) Application number: 93300465.7

(51) Int. Cl.⁵: **A01N 47/36**, A01N 25/22

(22) Date of filing: 22.01.93

③ Priority: 28.01.92 JP 53048/92 22:05.92 JP 174651/92

- (43) Date of publication of application : 04.08.93 Bulletin 93/31
- (A) Designated Contracting States:

 AT BE CH DE DK ES FR GB GR IE IT LI LU MC

 NL PT SE
- (1) Applicant: ISHIHARA SANGYO KAISHA, LTD. No. 3-22, Edobori 1-chome Nishi-ku Osaka (JP)
- 72 Inventor: Yoshida, Tsunezo, c/o Ishihara Sangyo K. K.
 Chuo Kenkyusho, 3-1, Nishishibukawa 2-chome
 Kusatsu-shi, Shiga-ken (JP)
 Inventor: Kuriyama, Yasuhide, c/o Ishihara Sangyo K. K.
 Chuo Kenkyusho, 3-1, Nishishibukawa 2-chome
 Kusatsu-shi, Shiga-ken (JP)
 Inventor: Kanbayashi, Shigehisa, c/o Ishihara Sangyo K. K.
 Chuo Kenkyusho, 3-1, Nishishibukawa 2-chome
 Kusatsu-shi, Shiga-ken (JP)
- (74) Representative: Pearce, Anthony Richmond MARKS & CLERK Alpha Tower Suffolk Street Queensway Birmingham B1 1TT (GB)
- (54) Chemically stabilized herbicidal oil-based suspension.
- 57 Disclosed is a chemically stabilized herbicidal oil-based suspension, comprising N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl-3-dimethylaminocarbonyl-2-pyridinesulfonamide and/or its salt as an effective herbicidal component, urea, a vegetable oil and/or mineral oil, a surfactant and, as desired, an additional herbicidal component, a thickener, a solvent and other adjuvants. The present invention is characterized in that urea is added to a herbicidal oil-based suspension to suppress decomposition of the effective herbicidal component and to provide a chemically stabilized herbicidal oil-based suspension.

EP 0 554 015 A1

The present invention relates to a chemically stabilized herbicidal oil-based suspension containing N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfonamide (hereinafter referred to as compound A) and/or its salt as an effective herbicidal component, which suppress the decomposition of compound A and/or its salt.

The present inventors found previously that pyridinesulfonamide series compounds including compound A and the salts thereof exhibit a very high herbicidal effect when they can be applied to a wide range of weeds including strongly harmful weeds, and filed European Patent Application No. 87300502.9 (or Laid-open European Patent Application No. 232067) on the basis of this finding. The inventors also found that a suspended composition comprising at least one compound selected from the pyridinesulfonamide series compounds and the salts thereof, a vegetable oil and a surfactant, which are mixed at a predetermined ratio, permits improving the herbicidal effect and decreasing the amount of the effective herbicidal component, and filed European Patent Application No. 88309772.7 (or Laid-open European Patent Application No. 313317).

On the other hand, various methods have been proposed to date for suppressing the decomposition of herbicidal sulfonylurea series compounds similar to compound A within preparations of agricultural chemicals so as to stabilize the formulations. For example, a stabilized aqueous composition containing a carboxylate or an inorganic salt is disclosed in Published Unexamined Japanese Patent Application No. 59-205305 (or Laidopen European Patent Application No. 124295). A stabilized granular composition containing calcium carbonate and, as required, sodium tripolyphosphate is disclosed in Published Unexamined Japanese Patent Application No. 62-84004. A stabilized solid formulations of agricultural chemicals containing a mineral-based carrier and a vegetable oil or a solvent having a high boiling point is disclosed in Published Unexamined Japanese Patent Application No. 63-23806. Further, various stabilized compositions of agricultural chemicals containing urea, thiourea or derivatives thereof are disclosed in Published Unexamined Japanese Patent Application No. 50-40739, Published Unexamined Japanese Patent Application No. 51-12930, Published Unexamined Japanese Patent Application No. 52-117422, Published Unexamined Japanese Patent Application No. 52-117422, Published Unexamined Japanese Patent Application No. 56-169606 (or Laid-open Belgian Patent Application No. 888634 or Laid-open British Patent Application No. 2074871), and Published Unexamined Japanese Patent Application No. 57-18605.

However, any of the publications exemplified above does not teach at all an idea of adding urea to a herbicidal oil-based suspension containing compound A and/or a salt thereof for chemically stabilizing it.

As a result of an extensive research on the measure for chemically stabilizing a herbicidal oil-based suspension containing compound A and/or its salt, the present inventors have found that compound A and/or its salt can be prevented from decomposition by adding urea to the suspension so as to chemically stabilize it, arriving at the present invention.

According to the present invention, there is provided a chemically stabilized herbicidal oil-based suspension, comprising compound A and/or its salt, urea, a vegetable oil and/or mineral oil, and a surfactant.

The present invention also provides a chemically stabilized herbicidal oil-based suspension, comprising compound A and/or its salt, urea, a vegetable oil and/or mineral oil, a surfactant, and at least one additional herbicidal component (hereinafter referred to as other specified herbicidal component(s)) selected from the group consisting of 2,4-dichlorophenoxyacetic acid (common name: 2,4-D), its alkyl ester and salt, 3,6-dichloro-2-methoxybenzoicacid (common name: dicamba) and its salt,

- 2-chloro-4-ethylamino-6-isopropylamino-s-triazine (common name: atrazine),
- 3-(1-methylethyl)-1H-2,1,3-benzothiadiazin-4(3H)-one-2,2-dioxide (common name: bentazone),
- 2-chloro-2',6'-diethyl-N-(methoxymethyl)acetanilide (common name: alachlor),
- 2-chloro-6'-ethyl-N-(2-methoxy-1-methylethyl)aceto-otoluidide (common name: metolachlor),
- 45 2-chloro-N-isopropylacetanilide (common name: propachlor), N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine (common name: pendimethalin),
 - 2-(3,5-dichlorophenyl)-2-(2,2,2-trichloroethyl)oxirane (common name: tridiphane), methyl
 - 2-[[[4,6-bis-(difluoromethoxy)-pyrimidin-2-yl]aminocarbonyl]aminosulfonyl]benzoate (common name: primisulfuron-methyl) and its salt,
 - 3,5-dibromo-4-hydroxybenzonitrile (common name: bromoxynil), its carboxylic acid ester and salt,
 - 5,7-dimethyl-N-(2,6-dichlorophenyl)-1,2,4-triazolo [1,5-a]pyrimidine-2-sulfonamide (D489: a compound described in Plant Physiology, 1990, Vol. 93, pages 962 to 966), 2-chloro-N-(ethoxymethyl)-2'-ethyl-6'-methylacetanilide (common name; acetochlor).
 - O-(6-chloro-3-phenyl-4-pyridazinyl) S- octylcarbonothioate (common name: pyridate),
 - 1-(4,6-dimethoxypyrimidin-2-yl)-3-(3-ethylsulfonyl-2-pyridylsulfonyl)urea (DPX-E9636: a compound described in Short Review of Herbicides & PGRs, 1991, page 94) and its salt, 2-[2-chloro-4-(methylsulfonyl) benzoyl]-1,3-cyclohexanedione (common name: sulcotrione) and its salt, methyl 3-chloro-5-(4,6-dimethoxypyrimidin-2-ylcarbamoylsulfamoyl)-1-methylpyrazole-4-carboxylate (NC-319: a compound described in BRIGHTON

5

20

25

30

35

40

CROP PROTECTION CONFERENCE-Weeds-1991, page 31) and its salt, and 3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea (common name: linuron).

In other words, in a herbicidal oil-based suspension comprising compound A and/or its salt as an effective herbicidal component, or in a herbicidal oil-based suspension comprising compound A and/or its salt as well as other specified herbicidal component(s) as an effective herbicidal component, the chemically stabilized herbicidal oil-based suspension characterized in that urea is added to said suspension.

The chemically stabilized herbicidal oil-based suspension of the present invention comprises compound A and/or its salt, and, as required, other specified herbicidal component(s), and, urea, a vegetable oil and/or mineral oil, a surfactant and, optionally, a thickener, a solvent and other adjuvants. These components are mixed uniformly, or optional components are mixed in advance, followed by adding the other components, so as to obtain the suspension of the present invention in the form of an oil-based suspension concentrate or a formulation for ultra low volume spraying. In preparing the suspension of the present invention, it is possible to employ wet-grinding, as required.

The salts of compound A contained in the suspension of the present invention include, for example, salts with alkali metals such as sodium and potassium, salts with alkaline earth metals such as magnesium and calcium, and salts with amines such as monomethylamine, dimethylamine and triethylamine. It is possible for compound A to be present together with a salt thereof.

The other specified herbicidal components include, for example, salts, alkyl esters and carboxylic acid esters. The salts include those similar to the salts of compound A described above, salts with amines such as diolamine and trolamine, and dimethylammonium salt. The alkyl esters include the esters with ethyl, butyl, heptyl, octyl, iso-octyl and butoxyethyl groups. Further, the carboxylic acid esters includes the esters with carboxylic acids such as butanoic acid, heptanoic acid and octanoic acid.

Among the additional herbicidal component, preferred are 2,4-D, its alkyl ester and salt, dicamba and its salt, bromoxynil, its carboxylic acid ester and salt, pyridate, DPX-E9636 and its salt, and sulcotrion and its salt.

The vegetable oil and mineral oil used in the suspension of the present invention include, for example, olive oil, kapok oil, castor oil, papaya oil, camellia oil, palm oil, sesame oil, corn oil, rice bran oil, peanut oil, cotton seed oil, soybean oil, rapeseed oil, linseed oil, tung oil, sunflower oil, safflower oil, and liquid paraffin. Particularly, it is desirable to use corn oil and rapeseed oil. These vegetable oils and mineral oils can be used in the form of a mixture, if necessary.

The surfactant used in the present invention includes, for example, salt of alkyl sulfonic acid, salt of alkyl-benzene sulfonic acid, salt of lignin sulfonic acid, polyoxyethyleneglycol alkylether, polyoxyethylene lauryl ether, polyoxyethylene alkyl ether, polyoxyethylene alkylaryl ether, polyoxyethylene fatty acid ester, polyoxyethylene fatty acid ester, polyoxyethylene styrylphenyl ether, salt of polyoxyethylene sorbitan fatty acid ester, polyoxyethylene styrylphenyl ether, salt of polyoxyethylene alkylaryl ether sulfate, salt of polyoxyethylene alkylaryl ether sulfate, salt of polyoxyethylene alkylaryl phosphoric acid ester, polyoxyethylene hydrogenated castor oil, salt of styrylphenyl phosphoric acid, condensate of naphthalenesulfonate with formalin, salt of benzoic acid, fatty acid polyglyceride, glycerin fatty acid ester, sorbitan monooleic acid ester, polyoxyethylene sorbitan monolauric acid ester, and fatty acid alcohol polyglycol ether. These surfactants can be used in the form of a mixture, if necessary.

The thickener which is contained as required in the chemically stabilized herbicidal oil-based suspension of the present invention includes, for example, silica and bentonite-alkylamino complex. Further, the solvent which is contained as required in chemically stabilized herbicidal oil-based suspension of the present invention includes, for example, aliphatic hydrocarbons such as normal paraffins and isoparaffins; aromatic hydrocarbons such as benzene, alkylbenzene, naphthalene, alkylnaphthalene, diphenyl, and phenyl xylyl ethane; heterocyclic compounds such as N-methylpyrrolidone and 1,3-dimethyl-2-imidazolidinone; alcohols; ethers; ketones; and esters.

The specific materials exemplified above as the thickener and solvent used in the present invention can be used in the form of a mixture, if necessary.

Concerning the mixing ratio of the components, the chemically stabilized herbicidal oil-based suspension of the present invention comprises, based on the total weight of the suspension, 0.5 to 20 parts by weight, preferably 1 to 6 parts by weight, more preferably 2 to 6 parts by weight, of compound A and/or its salt, 0.5 to 75 parts by weight, preferably 0.5 to 50 parts by weight of other specified herbicidal component(s) in the case where the suspension contains said another specified herbicidal component, 0.2 to 10 parts by weight, preferably 0.5 to 5 parts by weight, of urea, 19 to 93.8 parts by weight, preferably 30 to 88 parts by weight, more preferably 38.5 to 88 parts by weight of a vegetable oil and/or mineral oil, 5 to 25 parts by weight, preferably 8 to 15 parts by weight, of a surfactant, 0 to 5 parts by weight, preferably 0.5 to 3 parts by weight, more preferably 1 to 2 parts by weight, of a thickener when added to the suspension. Further, suitable amounts

5

10

20

25

30

35

40

45

of other adjuvants are also contained as required in the suspension of the present invention.

Exemplified below are formulations of the chemically stabilized herbicidal oil-based suspension of the present invention. Of course, the present invention is not restricted to the formulations exemplified below.

5 Formulation Example 1

		Parts by weight
	(1) Compound A (purity: 93.8%)	4.91
10	(2) A mixture of polyoxyethylene monylphenyl ether, dialkylsulfosuccinate, polyoxyethylene hydrogenated castor oil, and polyglycerol esters of fatty acid (trade name: Sorpol 3815K, manufactured by Toho Chemical Industry, Co., Ltd.)	12.55
15	(3) Bentonite-alkylamino complex (trade name: New D ORBEN, manufactured by Shiraishi Kogyo Kaisha, Ltd.)	2.09
	(4) Urea	1.05
	(5) Corn oil	79.40

20

A mixture of components (1) to (5) was subjected to wet-grinding for 15 minutes using a wet-grinding machine DYNO-MILL type KDL manufactured by Willy A. Bachofen. The wet-grinding machine was loaded at a loading rate of 60% with glass beads having a diameter of 1.0 mm and rotated at a peripheral speed of 10.5 m/sec and an oil-based suspension concentrate was obtained.

25

30

Formulation Example 2

An oil-based suspension concentrate was obtained as in Formulation Example 1, except that urea was used in an amount of 2.09 parts by weight in contrast to 1.05 parts by weight in Formulation Example 1, and that corn oil was used in an amount of 78.36 parts by weight in place of 79.40 parts by weight in Formulation Example 1.

Formulation Example 3

35

An oil-based suspension concentrate was obtained as in Formulation Example 1, except that urea was used in an amount of 3.14 parts by weight in contrast to 1.05 parts by weight in Formulation Example 1, and that corn oil was used in an amount of 77.31 parts by weight in place of 79.40 parts by weight in Formulation Example 1.

Formulation Example 4

An oil-based suspension concentrate was obtained as in Formulation Example 1, except that the wetgrinding was performed for 30 minutes in place of 15 minutes in Formulation Example 1.

45

50

Formulation Example 5

		Parts by weight
5	(1) Compound A (purity: 93.8%)	2.79
	(2) Octanoic acid ester of 3,5-dibromo-4-hydroxybenzonitrile (purity: 94.6%)	25.38
10	(3) A mixture of glycerine fatty acid ester and polyoxyethylene alkylaryl ether (trade name: GERONOL VO/278, manufactured by Rhone-Poulenc)	9.44
	(4) Fine amorphous silica (trade name: AEROSIL R974, manufactured by Degussa)	0.94
	(5) Urea	0.57
15	(6) Corn oil	60.88

A mixture of components (1) to (6) was subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and an oil-based suspension concentrate was obtained.

20 Formulation Example 6

25

30

35

40

An oil-based suspension concentrate was obtained as in Formulation Example 5, except that urea was used in an amount of 0.75 part by weight in place of 0.57 part by weight in Formulation Example 5, and that corn oil was used in an amount of 60.70 parts by weight in place of 60.88 parts by weight in formulation Example 5.

Formulation Example 7

An oil-based suspension concentrate was obtained as in Formulation Example 5, except that urea was used in an amount of 0.94 part by weight in place of 0.57 part by weight in Formulation Example 5, and that corn oil was used in an amount of 60.51 parts by weight in place of 60.88 parts by weight in Formulation Example 5.

Formulation Example 8

An oil-based suspension concentrate was obtained as in Formulation Example 5, except that urea was used in an amount of 1.42 parts by weight in place of 0.57 part by weight in Formulation Example 5, and that corn oil was used in an amount of 60.03 parts by weight in place of 60.88 parts by weight in Formulation Example 5.

Formulation Example 9

		Parts by weight
45	(1) Compound A (purity: 93.8%)	2.79
	(2) Octanoic acid ester of 3,5-dibromo-4-hydroxybenzonitrile (purity: 94.6%)	25.38
	(3) GERONOL VO/278 (trade name)	9.44
50	(4) AEROSIL R974 (trade name)	1.51
	(5) Urea	0.94
	(6) Aromatic solvent with a light boiling point (trade name: SOLVESSO 200, manufactured by Exxon Chemical Ltd.)	18.87
55	(7) Carn oil	41.07

A mixture of components (1) to (7) was subjected to wet-grinding for 15 minutes with the same wet-grinding

machine and conditions as in Formulation Example 1, and an oil-based suspension concentrate was obtained.

Formulation Example 10

An oil-based suspension concentrate was obtained as in Formulation Example 9, except that an aromatic solvent with a high boiling point (trade name: HISOL SAS-296, manufactured by Nippon Petrochemicals Co., Ltd.) was used in place of SOLVESSO 200 (trade name) used in Formulation Example 9.

Formulation Example 11

10

5

An oil-based suspension concentrate was obtained as in Formulation Example 9, except that wet-grinding was performed for 30 minutes in place of 15 minutes in Formulation Example 9.

Formulation Example 12

15

20

25

30

35

	Parts by weight
(1) Compound A (purity: 93.8%)	4.69
(2) Ethyl 2,4-dichlorophenoxyacetate (purity: 97.8%)	21.47
(3) GERONOL VO/278 (trade name)	10.00
(4) AEROSIL R974 (trade name)	1.00
(5) Urea	1.00
(6) Corn oil	61.84

A mixture of components (1) to (6) was subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and an oil-based suspension concentrate was obtained.

Formulation Example 13

An oil-based suspension concentrate was obtained as in Formulation Example 12, except that urea was used in an amount of 2.00 parts by weight in place of 1.00 part by weight used in Formulation Example 12, and that corn oil was used in an amount of 60.84 parts by weight in place of 61.84 parts by weight used in Formulation Example 12.

Formulation Example 14

Ю

An oil-based suspension concentrate is obtained as in Formulation Example 1, except that rapeseed oil is used in place of corn oil used in Formulation Example 1.

Formulation Example 15

50

45

An oil-based suspension concentrate is obtained as in Formulation Example 1, except that fatty alcohol polyglycol ether (trade name: Emulsogen EL-100, manufactured by Hoechst) is used in place of Sorpol 3815K (trade name) used in Formulation Example 1.

55

Formulation Example 16

		Parts by weight
5	(1) Compound A (purity: 93.8%)	2.80
	(2) Octanoic acid ester of 3,5-dibromo-4-hydroxybenzonitrile (purity: 94.2%)	25.49
	(3) GERONOL VO/278 (trade name)	11.32
10	(4) Bentonite-alkylamino complex (trade name: BENTONE SD-1, manufactured by RHEOX, Inc.)	2.36
	(5) HISOL SAS-296 (trade name)	18.87
15	(6) Urea	0.94
10	(7) Rapeseed oil	38.22

A mixture of components (1) to (7) was subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and an oil-based suspension concentrate was obtained.

Formulation Example 17

20

25

30

	Parts by weight
(1) Compound A (purity: 93.8%)	2.45
(2) Sorpol 3815K (trade name)	12.55
(3) New D ORBEN (trade name)	2.09
(4) Urea	1.05
(5) Corn oil	81.86

A mixture of components (1) to (5) is subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and a formulation for ultra low volume spraying is obtained.

Formulation Example 18

A formulation for ultra low volume spraying is obtained as in Formulation Example 17, except that compound A is used in an amount of 1.23 parts by weight in place of 2.45 parts by weight used in Formulation Example 17, and that corn oil is used in an amount of 83.08 parts by weight in place of 81.86 parts by weight used in Formulation Example 17.

50

45

Formulation Example 19

		Parts by weight
5	(1) Compound A (purity: 93.8%)	1.40
	(2) Octanoic acid ester of 3,5-dibromo-4-hydroxybenzonitrile (purity: 94.6%)	12.69
	(3) GERONOL VO/278 (trade name)	9.44
10	(4) AEROSIL R974 (trade name)	1.51
	(5) Urea	0.94
	(6) SOLVESSO 200 (trade name)	18.87
15	(7) Corn oil	55.15

A mixture of components (1) to (7) is subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and a formulation for ultra low volume spraying is obtained.

Comparative Formulation Example 1

	Parts by weight
(1) Compound A (purity: 93.8%)	4.91
(2) Sorpol 3815K (trade name)	12.55
(3) New D ORBEN (trade name)	2.09
(4) Corn oil	80.45

A mixture of components (1) to (4) was subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and an oil-based suspension concentrate was obtained.

Comparative Formulation Example 2

			Parts by weigh
40	(1)	Compound A (purity: 93.8%)	2.79
40	(2)	Octanoic acid ester of 3,5-dibr	omo-4-
	hydrox	<pre>sybenzonitrile (purity: 94.6%)</pre>	25.38
45	(3)	GERONOL VO/278 (trade name)	9.44
	(4)	AEROSIL R974 (trade name)	0.94
	(5)	Corn oil	61.45

A mixture of components (1) to (5) was subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and an oil-based suspension concentrate was obtained.

55

20

25

30

Comparative Formulation Example 3

	Parts by weight
(1) Compound A (purity: 93.8%)	4.69
(2) Ethyl 2,4-dichlorophenoxyacetate (purity: 97.8%)	21.47
(3) GERONOL VO/278 (trade name)	10.00
(4) AEROSIL R974 (trade name)	1.00
(5) Corn oil	62.84

A mixture of components (1) to (5) was subjected to wet-grinding for 15 minutes with the same wet-grinding machine and conditions as in Formulation Example 1, and an oil-based suspension concentrate was obtained. These formulations and comparative formulations were subjected to various tests as follows:

Test 1

50 m ℓ of the oil-based suspension concentrate obtained in each of Formulation Examples 1 to 3 and Comparative Formulation Example 1 was put in a glass container provided with stopper, and subjected to an accelerated storage stability test for one week under a constant temperature of 60°C. The rates of decomposition of compound A were determined with HPLC. The results are shown in Table 1.

25

5

10

15

20

30

35

Table 1

Formulation Example	Decomposition rate (%) of Compound A	
1	5.9	
2	5.6	
3	6.5	
Comparative Formulation Example 1	12.6	

As apparent from Table 1, the decomposition of compound A was markedly suppressed in each of Formulation Examples 1 to 3, compared with Comparative Formulation Example 1.

HPLC after predetermined period. The results are shown in Table 2.

50 ml of the oil-based suspension concentrate obtained in each of Formulation Examples 5 to 8 and Com-

Test 2

40 parative Formulation Example 2 was put in a glass container provided with stopper, and subjected to an accelerated storage stability test for one month under a constant temperature of 50°C and for 2 weeks under a constant temperature of 60°C, respectively. The rates of decomposition of compound A were determined with

45

50

Table 2

Formulation Example	Decomposition rate (%) of Compound A			
	50°C 60°C		>	
1. 1.	2 weeks later	1 month later	1 week later	2 weeks later
5	1.2	2.4	4.0	5.6
6	1.5	2.7	3.8	4.8
7	0.5	1.9	3.5	3.4
8	0.5	2.7	0	3.4
Comparative Formulation Example 2	2.7	7.3	11.0	21.0

Table 2 clearly shows that the decomposition of compound A was markedly suppressed in each of Formulation Examples 5 to 8, compared with Comparative Formulation Example 2. Further, decomposition of octanoic acid ester of 3,5-dibromo-4-hydroxybenzonitrile was not recognized in each of these Formulation Examples and Comparative Formulation Example.

Test 3

5

10

15

20

25

30

35

40

45

50

55

50 ml of the oil-based suspension concentrate obtained in each of Formulation Examples 9 and 10 was put in a glass container provided with stopper, and subjected to an accelerated storage stability test for one week under a constant temperature of 60°C. The rates of decomposition of compound A were determined with HPLC. The result are shown in Table 3.

Table 3

Formulation Example	Decomposition rate (%) of Compound A	
9		0
10		0.7

Table 3 shows that the decomposition of compound A was markedly suppressed in each of Formulation Examples 9 and 10, as in tests 1 and 2. Further, decomposition of octanoic acid ester of 3,5-dibromo-4-hydroxybenzonitrile was not recognized in each of these Formulation Examples.

Test 4

50 mℓ of the oil-based suspension concentrate obtained in each of Formulation Examples 12 and 13 and Comparative Formulation Example 3 was put in a glass container provided with stopper, and subjected to an accelerated storage stability test for one month under a constant temperature of 50°C and for one week under a constant temperature of 60°C, respectively. The rates of decomposition of compound A were determined with HPLC after predetermined period. The results are shown in Table 4.

Table 4

Formulation Example	Decomposition rate (%) of Compound A			
	50°	60°C		
	2 weeks later	1 month later	1 week later	
12	3.8	5.4	2.7	
13	1.6	2.1	. 0	
Comparative Formulation Example 3	2.9	7.0	12.0	

Table 4 clearly shows that the decomposition of compound A was markedly suppressed in each of Formulation Examples 12 and 13, compared with Comparative Formulation Example 3. Further, decomposition of ethyl 2,4-dichlorophenoxyacetate was not recognized in each of these Formulation Examples and Comparative Formulation Example.

Claims

5

10

- A chemically stabilized herbicidal oil-based suspension, comprising N-[(4,6-dimethoxypyrimidine-2-yl) aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfonamide and/or its salt, urea, a vegetable oil and/or mineral oil, and a surfactant.
- 2. In a herbicidal oil-based suspension comprising N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfonamide and/or its salt as an effective component, a chemically stabilized herbicidal oil-based suspension characterized in that urea is added to said suspension.
- 3. The chemically stabilized herbicidal oil-based suspension according to claim 1 or 2, characterized in that said N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfoneamide and/or its salt is N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfonamide.
- 4. The chemically stabilized herbicidal oil-based suspension according to claim 1, comprising 0.5 to 20 parts by weight of N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfonamide and/or its salt, 0.2 to 10 parts by weight of urea, 19 to 93.8 parts by weight of a vegetable oil and/or mineral oil, and 5 to 25 parts by weight of a surfactant.
- 5. The chemically stabilized herbicidal oil-based suspension according to claim 1, characterized by further comprising a thickener and/or a solvent.
 - 6. The chemically stabilized herbicidal oil-based suspension according to claim 1, characterized in that said vegetable oil and/or mineral oil is a vegetable oil.
- The chemically stabilized herbicidal oil-based suspension according to claim 6, characterized in that said vegetable oil is at least one vegetable oil selected from the group consisting of corn oil and rapeseed oil.
- 8. A chemically stabilized herbicidal oil-based suspension, comprising N-[(4,6-dimethoxypyrimidin-2-yl) amino-carbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfonamide and/or its salt, at least one additional herbicidal component, urea, a vegetable and/or mineral oil, and a surfactant, said additional herbicidal component being selected from the group consisting of 2,4-dichlorophenoxyacetic acid, its alkyl ester and salt, 3,6-dichloro-2-methoxybenzoic acid and its salt, 2-chloro-4-ethylamino-6-isopropylamino-s-triazine, 3-(1-methylethyl)-1H-2,1,3-benzothiadiazin-4(3H)-one-2,2-dioxide, 2-chloro-2',6'-diethyl-N-(methoxymethyl)acetanilide, 2-chloro-6'-ethyl-N-(2-methoxy-1-methylethyl)aceto-o-toluidide, 2-chloro-N-isopropylacetanilide, N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine,
- 2-(3,5-dichlorophenyl)-2-(2,2,2-trichloroethyl)oxirane, methyl 2-[[[4,6-bis-(difluoromethoxy)-pyrimidin-2-yl] aminocarbonyl]aminosulfonyl]benzoate and its salt,
 3,5-dibromo-4-hydroxybenzonitrile, its carboxylic acid ester and salt,

- 5,7-dimethyl-N-(2,6-dichlorophenyl)-1,2,4-triazolo [1,5-a]pyrimidine-2-sulfonamide,
- 2-chloro-N-(ethoxymethyl)-2'-ethyl-6'-methylacetanilide, O-(6-chloro-3-phenyl-4-pyridazinyl) S-octyl-carbonothioate,
- 1-(4,6-dimethoxypyrimidin-2-yl)-3-(3-ethylsulfonyl-2-pyridylsulfonyl)urea and its salt,
- 5 2-[2-chloro-4-(methylsulfonyl) benzoyl]-1,3-cyclohexanedione and its salt, methyl 3-chloro-5-(4,6-dimethoxypyrimidin-2-ylcarbamoylsulfamoyl)-1-methylpyrazole-4-carboxylate and its salt, and 3-(3,4-dichlorophenyl)-1-methylurea.
- 9. In a herbicidal oil-based suspension comprising N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfoneamide and/or its salt and at least one additional herbicidal component selected from the group consisting of 2,4-dichlorophenoxyacetic acid, its alkyl ester and salt,
 - 3,6-dichloro-2-methoxybenzoic acid and its salt,
 - 2-chloro-4-ethylamino-6-isopropylamino-s-triazine,
 - 3-(1-methylethyl)-1H-2,1,3-benzothiadiazin-4(3H)-one-2,2-dioxide,
- 2-chloro-2',6'-diethyl-N-(methoxymethyl)acetanilide,
 - 2-chloro-6'-ethyl-N-(2-methoxy-1-methylethyl)aceto-o-toluidide, 2-chloro-N-isopropylacetanilide,
 - N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine,
 - 2-(3,5-dichlorophenyl)-2-(2,2,2-trichloroethyl)oxirane, methyl 2-[[[4,6-bis-(difluoromethoxy)-pyrimidin-2-yl] aminocarbonyl]aminosulfonyl]benzoate and its salt,
- 3,5-dibromo-4-hydroxybenzonitrile, its carboxylic acid ester and salt,
 - 5,7-dimethyl-N-(2,6-dichlorophenyl)-1,2,4-triazolo [1,5-a]pyrimidine-2-sulfonamide,
 - 2-chloro-N-(ethoxymethyl)-2'-ethyl-6'-methylacetanilide, O-(6-chloro-3-phenyl-4-pyridazinyl) S-octyl-carbonothioate.
 - 1-(4,6-dimethoxypyrimidin-2-yl)-3-(3-ethylsulfonyl-2-pyridylsulfonyl)urea and its salt,
- 2-[2-chloro-4-(methylsulfonyl) benzoyl]-1,3-cyclohexanedione and its salt, methyl 3-chloro-5-(4,6-dimethoxypyrimidin-2-yl-carbamoylsulfamoyl)-1-methylpyrazole-4-carboxylate and its salt, and 3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea which are effective components, a chemically stabilized herbicidal oil-based suspension characterized in that urea is added to said suspension.
- 10. The chemically stabilized herbicidal oil-based suspension according to claim 8, comprising 0.5 to 20 parts by weight of N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl]-3-dimethylaminocarbonyl-2-pyridinesulfonamide and/or its salt, 0.5 to 75 parts by weight of at least one additional herbicidal component, 0.2 to 10 parts by weight of urea, 19 to 93.8 parts by weight of a vegetable oil and/or mineral oil, and 5 to 25 parts by weight of a surfactant.
- 11. The chemically stabilized herbicidal oil-based suspension according to claim 8 or 9, characterized in that said additional herbicidal component is selected from the group consisting of 2,4-dichlorophenoxyacetic acid, its alkyl ester and salt, 3,6-dichloro-2-methoxy benzoic acid and its salt,
 - 3,5-dibromo-4-hydroxybenzonitrile, its carboxylic acid ester and salt, O-(6-chloro-3-phenyl-4-pyridazinyl) S-octylcarbonothioate,
 - 1-(4,6-dimethoxypyrimidine-2-yl)-3-(3-ethylsulfonyl-2-pyridylsulfonyl) urea and its salt, and
 - 2-[2-chloro-4-(methylsulfonyl) benzoyl]-1,3-cyclohexanedione and its salt.
 - 12. The chemically stabilized herbicidal oil-based suspension according to claim 8 or 9, characterized in that said additional herbicidal component is selected from the group consisting of 2,4-dichlorophenoxyacetic acid, its alkyl ester and salt, and
 - 3,5-dibromo-4-hydroxybenzonitrile, its carboxylic acid ester and salt.
 - 13. An use of urea to suppress decomposition of N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl]-3-dimethy-laminocarbonyl-2-pyridinesulfonamide and/or its salt contained as an effective component in a chemically stabilized herbicidal oil-based suspension.
 - 14. A method of using urea to suppress decomposition of N-[(4,6-dimethoxypyrimidin-2-yl) aminocarbonyl]3-dimethylaminocarbonyl-2-pyridinesulfonamide and/or its salt contained as an effective component in a chemically stabilized herbicidal oil-based suspension.

55

40

45



EUROPEAN SEARCH REPORT

Application Number

EP 93 30 0465

ategory	Official Control of the form		Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
۱,D	EP-A-0 313 317 (ISH			A01N47/36 A01N25/22
	DD-A-252 748 (VEB F/	AHLBERG-LIST)		,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,
				TECHNICAL FIELDS
	·			SEARCHED (Int. Cl.5)
				AOIN
		·		
		·		
			·	
	The present search report has b	cen drawn up for all claims Date of completion of the search		Domino
	THE HAGUE	29 APRIL 1993		DECORTE D.M.
X : pa	CATEGORY OF CITED DOCUME rticularly relevant if taken alone rticularly relevant if combined with an	E : earlier patent after the filing other D : 40ccument cite	document, but put g date ed in the application	biished on, or on
ďα	cument of the same category chnological background	L : document cite	d for other reason	5

THIS PAGE BLANK (USPTO)